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Letter

Competing structures in the beyond neutron N = 104 midshell nucleus ^{184}Pb

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ABSTRACT

Proton-neutron configurations in atomic nuclei can manifest in a variety of different structures, and exotic nuclei provide fruitful ground for exploring their relation to fundamental symmetries. Here, we report on in-beam γ-ray spectroscopy of the very neutron-deficient ^{184}Pb nucleus using the JUROGAM II spectrometer at the RITU separator in a recoil-decay tagging experiment. For the first time, transitions from the non-yrast states have been observed in a Pb nucleus beyond the neutron $N = 104$ midshell. In comparison to neighboring Pb isotopes and theoretical models, de-excitation patterns from these states are similar to those associated with predominantly oblate shapes in heavier Pb isotopes. Together with the known predominantly prolate yrast band and the spherical ground state, this suggests that triple-shape coexistence prevails in the ^{184}Pb nucleus.

1. Introduction

The interplay of proton-neutron configurations in atomic nuclei gives rise to diverse phenomena that are not present in other domains of physics [1]. In the neutron-deficient Pb region, the competing structures are often linked to multiparticle-multihole proton excitations across the closed $Z = 82$ shell [2,3]. These structures intrude close to the spherical ground states as the neutron midshell at $N = 104$ is approached. Mean-field methods offer a complementary perspective, associating each intruder minimum with a distinct collective shape. Early calculations of quadrupole potential energy surfaces within the Strutinsky approach [4,5], and subsequent self-consistent mean-field approaches using Skyrme and Gogny interactions, have confirmed the presence of spherical ground states with low-lying oblate and prolate minima [6–11,13]. Regarding ^{184}Pb, these calculations predict the prolate minimum to be lower in energy than the oblate one. This contrasts to the finite-range liquid-drop model calculations that predict the coexistence of different shapes, but with the oblate minimum being lower in energy than the prolate one [12]. While the coexisting deformed minima have been produced in several theoretical calculations, very few of them report on the characteristics of states with $J^{\pi} > 0^+$, or transitions from them, for nuclei beyond the $N = 104$ neutron midshell [2] [9,11,13].

Experimental efforts to investigate neutron-deficient Pb nuclei have been extensive, yet the information remains far from complete. While the charge-radii measurements of these nuclei have confirmed the sphericity of their ground states [14], the low-lying 0^+ states have been investigated through the decay of parental nuclei, as demonstrated in the α-decay fine-structure measurements of Po nuclei [15–18] and the β-decay studies of Bi nuclei [3,19]. In-beam γ-ray spectroscopic methods, employing fusion-evaporation reactions, have been pivotal in studies of bands built on top of the 0^+ states [20,21]. The transition energies and probabilities obtained for those bands exhibit characteristic features of rotating nuclei, suggesting deformed intrinsic shapes [22–31], and direct evidence for the existence of a prolate minimum was obtained through the particle-core coupling in ^{185}Pb [32]. Recently, simultaneous in-beam γ-ray and electron spectroscopy experiments have provided invaluable information for linking the 0^+ band-head states with corresponding bands, and for the configuration mixing via $E0$ transitions [31,33,34].

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1 Deceased.
2 I.e. neutron numbers $N < 104$.

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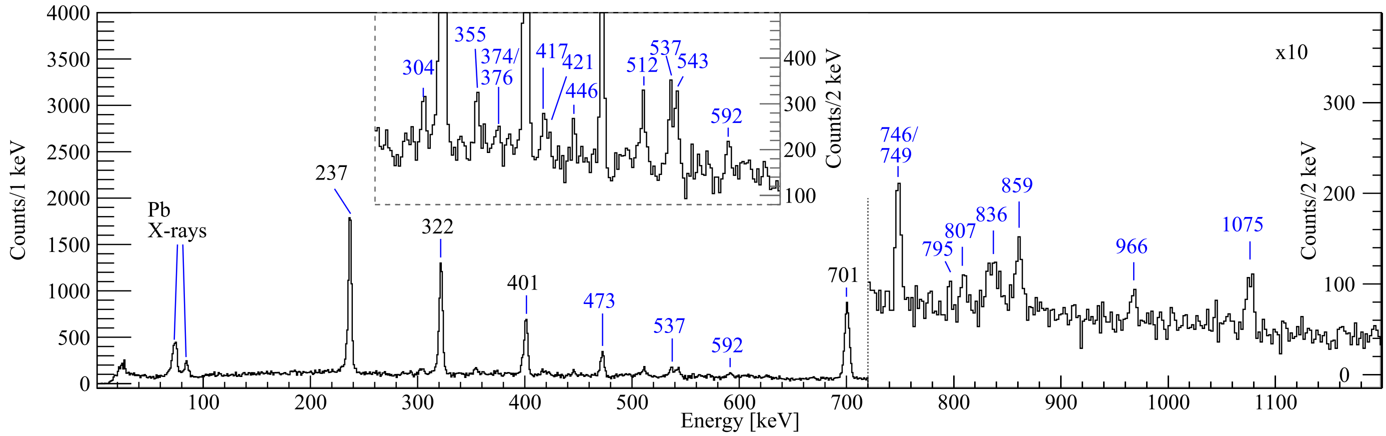

Fig. 1. Recoil-gated, α-tagged γ-ray energy spectrum. The y-axis of the high energy section has been expanded by a factor of ten for better visualization. Close-up of the energy region from 260 to 640 keV is shown in the inset. The most prominent peaks are marked with transition energies and the new transitions in blue.

Excited states in ^{184}Pb were first observed in in-beam γ-ray spectroscopy measurements by Cocks et al. [27]. Through comparison with heavier Pb isotopes, they assigned the four observed transitions as part of a predominantly prolate rotational band. Soon after, the excited 0^+ state at 570(30) keV was discovered by Andreyev et al. [15] and later confirmed by Van de Vel et al. [18] in the α-decay of ^{188}Po. In this Letter, we have identified several new transitions in ^{184}Pb. In addition to extending the yrast band up to the $J^\pi = (14^+)$ state, we present the first observation of transitions from the non-yrast states in a Pb nucleus beyond the neutron $N = 104$ midshell. These findings facilitate the assessment of the evolution of the oblate minimum, which is predicted to rise in excitation energy in the Pb isotopes lighter than $A = 186$ [5].

2. Experimental details

The experiment was performed at the Accelerator Laboratory of the University of Jyväskylä, Finland. Nuclei of interest were synthesized via the ^{104}Pd(^{83}Kr,3n)^{184}Pb fusion-evaporation reaction. The ^{83}Kr^{16+} beam was produced in an ECR ion-source [35] and accelerated to an energy of 367 MeV employing the K130 cyclotron [36]. The beam, impinging on a self-supporting metallic foil target of ^{104}Pd with a thickness of 1 mg/cm^2 and enrichment greater than 98%, had an average current of 16 pnA.

Prompt γ rays were observed with the JUROGAM II spectrometer [37], which consisted of 15 EUROGAM Phase I and 24 EUROGAM Clover type Compton-suppressed germanium-detector modules installed at the target position. The Recoil Ion Transport Unit (RITU) [38,39], a gas-filled separator, was employed to separate fusion-evaporation residues (recoils in this work) from the primary and scattered beam. The GREAT spectrometer [40], located at focal plane of RITU, was used to identify recoils implanted in the double-sided silicon strip detector (DSSSD) based on their characteristic α-decay energy. The maximum search time between the recoil implantation and the ^{184}Pb α decay was 1.5 s, which is approximately three times the half-life of ^{184}Pb ($t_{1/2} = 490(250)$ ms) [41]. A box of PIN diodes, upstream from the DSSSD, was used to detect α-particles that escaped from the implantation detector. Moreover, three Clover type germanium detectors were installed around the DSSSD for the search of delayed transitions de-exciting via γ ray emission.

The beam was on target for a total of 174 hours, during which 1.5×10^5 α-particles assigned with the decay of ^{184}Pb were detected. Consequently, the estimated reaction cross-section was approximately 3.0 μb. Spectroscopic studies at this level are only feasible if the nucleus of interest can be unambiguously identified, and its prompt de-excitation can be selected from orders of magnitude higher amount of background events. The recoil-decay tagging (RDT) method exploited in the present work is an ideal technique to conduct these studies [42,43].

Data were collected using the fully digital total data read-out data acquisition system, where signals from each channel are collected without a common hardware trigger and events are time-stamped using a global 100 MHz metronome [44]. The time correlation of events was performed using the Grain software package [45] and the analysis was completed using the ROOT framework [46].

3. Data analysis

The power of RDT allowed for the assignment of a number of transitions to ^{184}Pb. This is demonstrated in Fig. 1, where an RDT γ-ray singles energy spectrum, free of contaminants arising from other open fusion-evaporation channels, is presented. In addition to known transitions, many new transitions have been identified and marked with blue labels. The spectrum shown in the inset and the high-energy region are presented with a more sensitive y-scale for better visualization of the less intense transitions. Unfortunately, statistics was insufficient to conduct angular or directional correlation analysis. Based on sum-energy arguments, intensity balances and $\gamma\gamma$ coincidence relations, a partial level scheme shown in Fig. 2 has been deduced. All spin and parity assignments are tentative as indicated in Fig. 2. Transitions, and related properties, assigned to ^{184}Pb are listed in Table 1.

Regardless of the very small production cross section, sufficient $\gamma\gamma$ coincidence statistics to aid construction of the partial level scheme was obtained for several transitions. Four sample spectra with gates on chosen transitions are presented in different panels in Fig. 3. In panel a), the sum of gates on the 401, 473 and 537 keV γ-rays, stemming from the highest known yrast-band $8_1^+ \rightarrow 6_1^+$ transition [27] and the proposed two preceding $10_1^+ \rightarrow 8_1^+$ and $12_1^+ \rightarrow 10_1^+$ transitions, respectively, are shown. The present peaks support the earlier assignment for the yrast band sequence but also allow for the extension of the band by three new transitions, reaching the state with spin and parity of $J^\pi = (14^+)$. It is noteworthy that no other transitions assigned to ^{184}Pb are prominent in the spectrum. Transitions not belonging to the yrast band are observed in panel b), where a coincidence gate on the 237 keV $4_1^+ \rightarrow 2_1^+$ transition has been applied. In addition to the yrast-band transitions, prominent peaks at 355, 512 and 543 keV are evident. When gating on the 322 keV $6_1^+ \rightarrow 4_1^+$ transition as demonstrated in panel c), only the peak at 543 keV of these three peaks remains. This provides strong support for assigning the 512 and 543 keV peaks to transitions de-exciting non-yrast states at 1450 and 1804 keV, respectively, and the 355 keV peak to a transition between these new states. Further evidence for this assignment is presented in panel d), where the coincidence gate on the 355 keV transition is shown. Transitions stemming from the de-excitation path via the 512 keV transition are clearly present and have been marked in the spectrum. Of particular interest are the peaks at 376 and 749 keV, which together with the 1075 and 701 keV transitions observed in the RDT

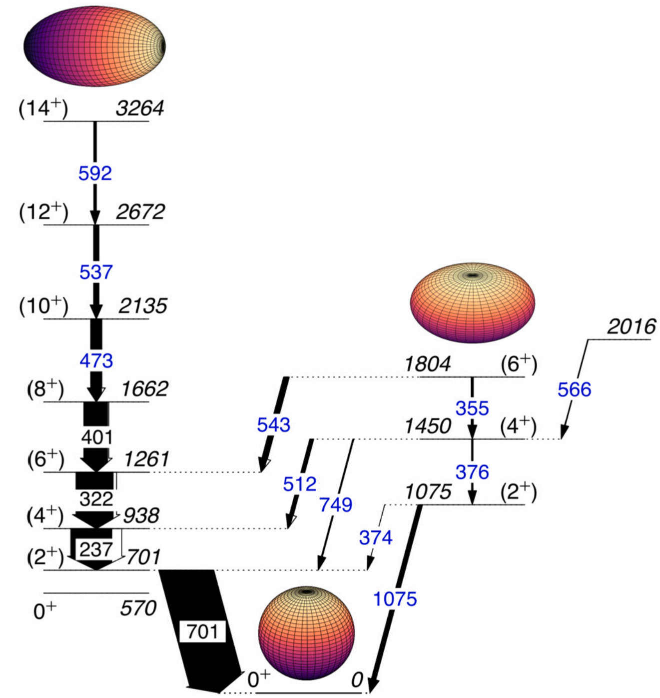

Fig. 2. Partial level scheme of ^{184}Pb obtained in the present work. The widths of the arrows are proportional to the total transition intensities and energies of new transitions are marked in blue. Proposed predominant shapes have been preliminary assigned on top of different structures.

Table 1
Transition properties obtained in the present work. For each transition, γ-ray energy (E_γ) and intensity (I_γ), energy of the initial state (E_i) and tentative spin and parity assignment of initial and final state ($J^\pi_i \to J^\pi_f$) has been listed.

E_γ [keV]	I_γ	E_i [keV]	$J^\pi_i \to J^\pi_f$
237.4(6)	779(22)	938.5(10)	$4^+_1 \to 2^+_1$
304.4(7)	38(8)	-	-
322.2(6)	707(22)	1260.9(11)	$6^+_1 \to 4^+_1$
354.8(6)	36(8)	1804.5(11)	$6^+_2 \to 4^+_2$
373.9(11)	4(3)*	1074.7(10)	$2^+_2 \to 2^+_1$
375.5(12)	25(18)*	1449.9(10)	$4^+_2 \to 2^+_2$
401.4(6)	451(20)	1662.3(13)	$8^+_1 \to 6^+_1$
416.9(6)	26(7)	-	-
421.4(9)	33(17)	-	-
446.1(6)	27(5)	-	-
472.8(6)	205(14)	2135.1(14)	$10^+_1 \to 8^+_1$
511.7(6)	66(16)	1449.9(10)	$4^+_2 \to 4^+_1$
537.0(6)	98(17)	2672.1(15)	$12^+_1 \to 10^+_1$
543.3(6)	97(13)	1804.5(11)	$6^+_2 \to 6^+_1$
566.0(9)	≤ 11	2015.9(14)	-
591.7(7)	47(11)	3263.8(17)	$14^+_1 \to 12^+_1$
701.1(6)	1000(27)	701.1(9)	$2^+_1 \to 0^+_1$
745.5(10)	74(11)	-	-
748.7(17)	21(11)	1449.9(10)	$4^+_2 \to 2^+_1$
794.8(8)	13(5)	-	-
806.5(8)	14(5)	-	-
835.5(18)	57(7)	-	-
859.3(7)	69(23)	-	-
966.1(8)	35(9)	-	-
1074.6(15)	84(11)	1074.7(10)	$2^+_2 \to 0^+_1$

* From singles data, an intensity of 29(18) was obtained for the 374/376 keV doublet. Individual transition intensities are based on the intensity ratio of I(376)/I(374) = 0.16(7) determined from $\gamma\gamma$ coincidence data by gating on the 374/376 keV doublet.

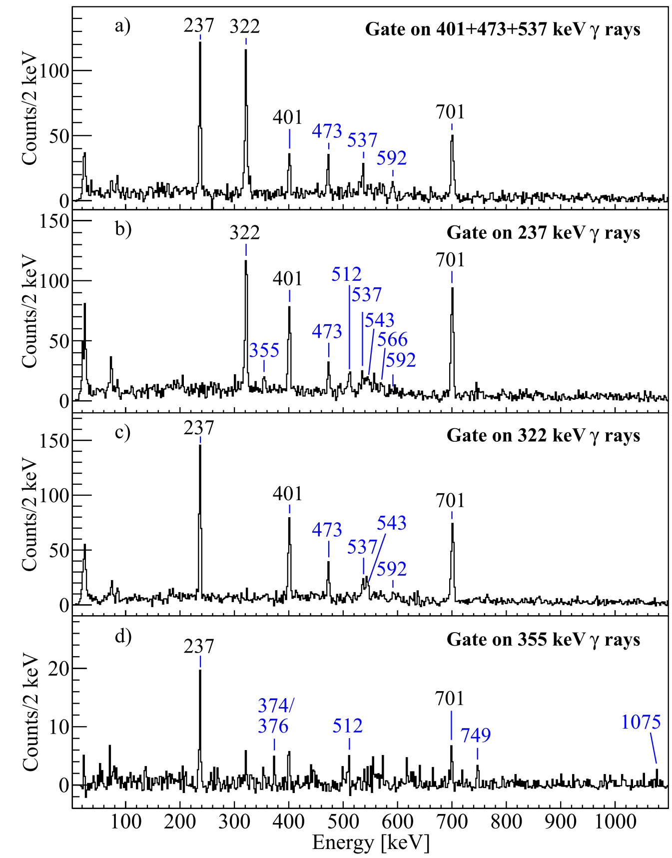

Fig. 3. Recoil-gated, α-tagged γ-ray energy spectra with gates on selected transitions. γ rays in coincidence with 401, 237, 322 and 355 keV transitions are shown in panels a), b), c) and d), respectively. The peaks assigned to ^{184}Pb are labeled with transition energies and the new transitions in blue. See text for more details.

γ-ray singles spectrum in Fig. 1, sum up, within the stated uncertainties, to 1450 keV, respectively. Therefore, we assign the peaks at 376 and 749 keV to de-excitations of the 1450 keV state, where the 749 keV transition feeds the 2^+_1 state at 701 keV and the 376 keV transition feeds a new state at 1075 keV. Moreover, $\gamma\gamma$ coincidence data indicate that the peak at ∼376 keV is a self-contained doublet, which also fits well in the level scheme as a transition between the 701 and 1075 keV states.

According to intensity balances when gating on γ rays corresponding to the 374/376 keV doublet, the 374 keV $2^+_2 \to 2^+_1$ transition lacks intensity. This could be explained by the presence of an $E0$ component, similar to that observed in ^{186}Pb and ^{188}Pb [34,33]. Missing intensity analysis can also be used for assessing transition multipolarities of the yrast-band transitions where the initial state for the transition of interest have only one de-excitation path. In the present work, $\gamma\gamma$ coincidence statistics were sufficient to extract information for the 237 keV $4^+_1 \to 2^+_1$ and 322 keV $6^+_1 \to 4^+_1$ transitions, while the 701 keV $2^+_1 \to 0^+_1$ transition was used for normalization. Based on data presented in Fig. 3a, $\alpha_{tot}(4^+_1 \to 2^+_1) = 0.13(21)$ and $\alpha_{tot}(6^+_1 \to 4^+_1) = 0.02(20)$ were obtained. These results support the tentative $E2$ multipolarity assignments, although they do not rule out $E1$ multipolarities. Calculated values for the $E2$ [$E1$] transitions are 0.24 [0.051] and 0.094 [0.025], respectively [47].

Peaks corresponding to the 446 and 966 keV transitions are in mutual coincidence. Similarly, the peaks at 304, 417, 421, 606, 746, 795, 807, 836 and 859 keV listed in Table 1 have been firmly assigned to ^{184}Pb, but solid evidence for placing them in the level scheme could not be found. It is noteworthy that these transitions may be located above isomeric states. The absence of delayed transitions assigned to ^{184}Pb at the focal plane suggests that potential isomeric states de-excite as the re-

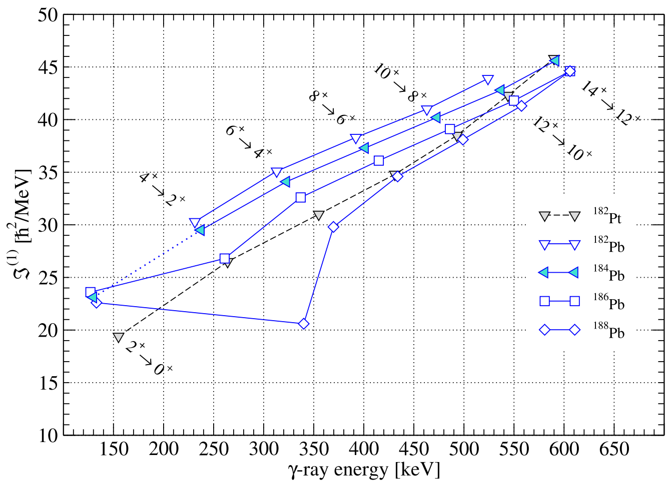

Fig. 4. Kinematic moment of inertia $\mathcal{J}^{(1)}$ as a function of γ-ray energy for the prolate bands in the neutron-deficient $^{182-188}$Pb nuclei [27,28,34,33]. A curve for the yrast band in ^{182}Pt is shown as a reference for an unperturbed prolate band in the region [48]. Dotted extension to the ^{184}Pb curve assumes the excited 0_2^+ state at 570 keV is the head of the predominantly prolate band.

coils pass through the RITU separator, indicating lifetimes ranging from a few to tens of nanoseconds. This is somewhat shorter than those observed in heavier Pb isotopes, but corresponds well with the expected level energy systematics, which implies that the transition from the isomeric state has higher energy in ^{184}Pb than in heavier Pb nuclei.

4. Discussion

In comparison to similar bands in the neighboring nuclei, the yrast-band in ^{184}Pb was associated as a rotational band with predominantly prolate shape by Cocks et al. [27]. These similarities are evident e.g. in kinematic moment of inertia plots. This is demonstrated in Fig. 4, where predominantly prolate bands in the ^{182,184,186,188}Pb isotopes together with an unmixed prolate band in the ^{182}Pt nucleus are plotted. A few interesting conclusions can be drawn from these curves. First, the extension of the band follows the linear trend, and there is no indication of a backbending phenomenon occurring up to the $J^\pi = (14^+)$ state. This covers rotational frequencies $\hbar\omega$ up to approximately 0.3 MeV and corresponds to a transition energy of 600 keV. As discussed in Ref. [26], alignment of a $\nu(i_{13/2})^2$ neutron pair would result in a smooth upbend that starts at lower rotational frequencies, similar to that observed for ^{182}Pt. A proton pair alignment is expected to be sharper and to occur at higher frequencies, matching better what is observed for ^{184}Pb. It is noteworthy that, based on the development of transition intensities along the yrast band and the present sensitivity, the observation of the $16_1^+ \rightarrow 14_1^+$ transition could be expected. The non-observation might be due to the reduced intensity of the $16_1^+ \rightarrow 14_1^+$ transition caused by backbending starting at $J^\pi = (16^+)$. Second, the smooth behavior of the yrast rotational band indicates little configuration mixing at low spin, in contrast to the kinks observed in the heavier ^{186,188}Pb isotopes. Finally, while the 131 keV $2_1^+ \rightarrow 0_2^+$ transition has not been observed due to the much more probable 701 keV $E2$ transition to the ground state, the assignment of the 0_2^+ state as the prolate band head is supported by the extension to the kinematic moment of inertia curve (see the dotted line in Fig. 4).

The state at 1075 keV de-excites via two different branches: one to the 0_1^+ ground state and another to the first excited 2_1^+ state. A spin $J = 1$ assignment would imply a highly non-yrast state, unlikely to be fed as strongly as observed in the present work. Conversely, with a $J = 3$ assignment, a dipole transition to the 2_1^+ state would be more probable than an octupole transition to the ground state, which is not what was observed. Similar arguments are valid for ruling out the odd-spin assignments for the other non-yrast states. Consequently, we assign states at 1075, 1450 and 1805 keV with tentative spins and parities of $2_2^+, 4_2^+$ and 6_2^+, respectively. These assignments follow the parabolic trend of the non-yrast even-spin states (see Fig. 8 of Ref. [34]), associated with predominantly oblate shape, near the $N = 104$ midshell Pb nuclei.

Table 2
Relative $B(E2)$ values extracted in the present work compared to those obtained for ^{186}Pb and ^{188}Pb. $M1$ admixtures are excluded from the $\Delta J = 0$ transitions, as the Clebsch–Gordan coefficients vanish when both bands have $K = 0$. The values for transitions from the non-yrast band have been normalized to the one with the highest transition strength.

J_i^π	J_f^π	E_γ [keV]	$B(E2)_{^{184}\mathrm{Pb}}$	$B(E2)_{^{186}\mathrm{Pb}}$	$B(E2)_{^{188}\mathrm{Pb}}$
4_2^+	4_1^+	511.7(6)	23(10)	34(5)	100(12)
	2_1^+	748.7(17)	3.2(17)	3.1(5)	9.5(1.1)
	2_2^+	375.5(12)	100(28)	100(14)	95(10)
2_2^+	2_1^+	373.9(11)	100(30)	≤ 86	100(11)
	0_3^+	-	-	≤ 100	-
	0_2^+	-	-	≤ 18	3.1(12)
	0_1^+	1074.6(15)	1.2(6)	3.6(5)	0.8(1)

The nature of the 2_2^+ and 4_2^+ non-yrast states can be assessed using the relative $B(E2)$ values extracted from intensity balances by gating on the feeding transitions, see Table 2. The vibrational character of the non-yrast band can be ruled out by the inter-band transition probability ratio $B(E2; 4_2^+ \rightarrow 4_1^+)/B(E2; 4_2^+ \rightarrow 2_1^+) = 7(5)$, which, according to the Alaga rules, should be close to unity [49]. In contrast, transitions between states with the same spin and parity appear to be strong. Table 2 also presents a comparison to corresponding values in ^{186}Pb and ^{188}Pb, where the sequences of non-yrast states have been associated with rotational, predominantly oblate bands. The relative $B(E2)$ values extracted for ^{184}Pb show a similar pattern to those in ^{186}Pb and ^{188}Pb, providing further support for an oblate assignment. The relatively high transition probability between states with the same spin and parity suggests the presence of prolate-oblate configuration mixing. In the case of 4^+ states, the relative $B(E2; 4_2^+ \rightarrow 4_1^+)$ value is smaller than that in the ^{188}Pb nucleus, suggesting a drop in the amount of mixing. This could also be expected from the level energy difference between the yrast and non-yrast 4^+ states, which increases from ^{188}Pb to ^{184}Pb. A similar interpretation can be drawn from the kinematic moments of inertia curves, where configuration mixing appears as deviations from the linear trend. In Fig. 4, this is most pronounced for the $4^+ \rightarrow 2^+$ transitions, for which the deviation decreases with decreasing neutron number.

5. Summary

In order to study the competing structures in neutron-deficient Pb nuclei beyond the $N = 104$ midshell, we performed an in-beam γ-ray spectroscopy experiment on the ^{184}Pb nucleus, employing the recoil-decay tagging technique. The results obtained allowed for the extension of the rotational yrast band by three new states and the discovery of de-excitation paths from the non-yrast states. Based on a comparison with the ^{186}Pb and ^{188}Pb isotopes, these new states are tentatively associated with structures built on the oblate minimum.

The reported data present spectroscopy of non-yrast states at the level of tens of nanobarns in cross-section, which is at the limits of current experiment reach. While experiments employing post-accelerated radioactive ion beams need a development step in intensity and purity for the beam [50,51], a relativistic Coulomb excitation experiment of ^{184}Pb following fragmentation also requires technical advances. Similarly, exploiting transfer reactions for these studies lacks feasible beam-target combinations to provide the required yields. To confirm the findings of the present work, we call for an in-beam spectroscopy experiment using a more powerful Ge-detector array, i.e., an array with higher efficiency without compromising granularity, in conjunction with a recoil separator. These data also invite theoretical calculations to better align with experimental findings.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

The data obtained in the present work and the corresponding metadata are available and open after an embargo period.

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